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Report No. IITRI-C6018-8
(Quarterly Report)

INVESTIGATION OF LIGHT SCATTERING
IN HIGHLY REFLECTING PIGMENTED COATINGS

National Aeronautics
and Space Administration

UNPUBLISHED PRELIMINARY DATA

Report No. IITRI-C6018-8
(Quarterly Report)

INVESTIGATION OF LIGHT SCATTERING IN HIGHLY REFLECTING PIGMENTED COATINGS

January 1, 1964 to April 1, 1964

Contract No. NASr-65(07)
IITRI Project C6018

Prepared by

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Office of Advanced Research and Technology
Washington, D. C.

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FOREWORD

This is Report No. IITRI-C6018-8 (Quarterly Report) of Project C6018, Contract No. NASr-65(07), entitled "Investigation of Light Scattering in Highly Reflecting Pigmented Coatings." This report covers the period from January 1, 1964 to April 1, 1964.

Major contributors to the program include Gene A. Zerlaut (Project Leader), Dr. S. Katz and Dr. B. Kaye (theoretical analyses), V. Raziunas (principle investigator), and Mrs. J. Allen (silver halide preparations).

Data are recorded in Logbooks C14085 and C13906.

Respectfully submitted,

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ABSTRACT

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The preparation and measurement of the optical properties of both mono- and bimodal dispersions of silver bromide in liquid and gelatin suspensions are reported. Scattering extrema as manifested in transmittance spectra were measured. The scattering extrema were determined as a function of gelatin film thickness. The transmittance of mixtures of two size distributions -- to give a bimodal particle distribution -- were determined. Multipole interactions -- resulting in fine structure in the transmittance measurements -- were observed on liquid suspensions of 1.3μ polystyrene spheres. These observations have not been reported in the literature.

Author

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I. INTRODUCTION

The aim of this program is the definition of the light-scattering parameters which are necessary for the maximum reflection of solar radiation. The definition of these factors should facilitate the eventual development of more highly efficient solar reflectors and, perhaps more important, should serve to extend out ability to apply light-scattering theory to the solution of other problems.

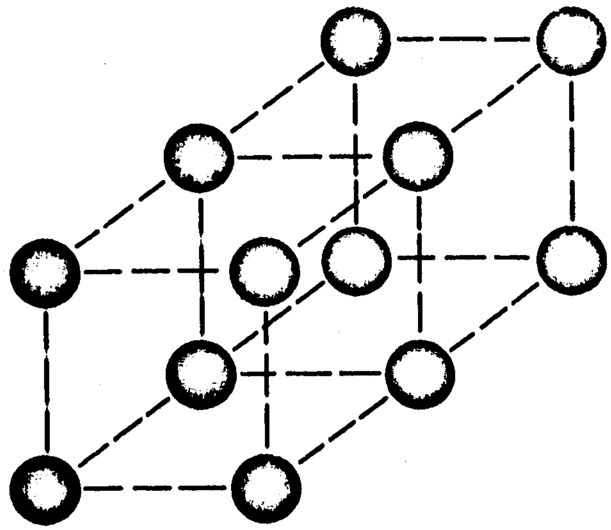
Previous quarterly reports have dealt with the problem of preparation of arrays of closely sized silver chloride particles and the measurement of their optical properties. Particular attention was devoted to the measurement of size distributions and concentrations of both liquid and gelatin suspensions and to the measurements of the transmittance, absorbance and radial scattering of gelatin suspensions. A general review of applicable light scattering theory has also been provided.

This report deals with the preparation and measurement of the optical properties of both mono- and bimodal dispersions of liquid and gelatin silver bromide particles. The work reported herein involved the use of silver bromide due to the discovery that better control of particle size and quality could be achieved compared to silver chloride. Of significance has been the determination of scattering extrema as manifested in the transmittance curves. Also of importance -- especially insofar as it represents observations not reported in the literature -- is the appearance of multiple interactions in transmittance measurements which were made on liquid suspenisons of 1.3 μ polystyrene spheres.

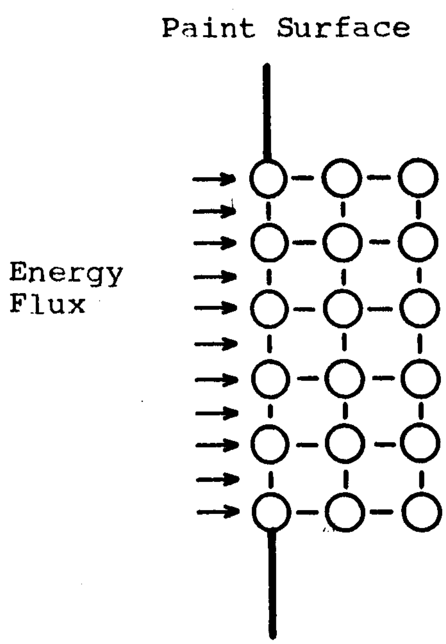
II. LIGHT-SCATTERING THEORY AND MULTIPLE SCATTERING SYSTEMS

A principal objective of the research program is the application of single particle scattering theory to particle arrays in an attempt to explain the scattering behavior of polydisperse pigmented coatings. The first problem is what constitutes an adequate model for studying the penetration of radiation through the two-phase system of the pigment and the matrix. The simplest structural model is a monosized pigment in regular array; i.e., all particle centers are located at the points on a cubic lattice. This type of array is shown schematically in Figure 1. The energy flux traversing an array normal to the surface could probably be studied by using diffraction theory.

The regular array appears to be the simplest model for studying multiple interaction. However, it has the disadvantage that the density of scattering centers is not independent of the direction of the incident radiation. In fact, a random array of particles may be a simpler model to treat since the properties of the array on average (averaged over a sufficinet distance) will be independent of the direction of travel. The only effect of non-normal incidence is that a given thickness of film appears thicker. These effects are illustrated in Figure 2. It may therefore be possible to study several random walks through an array and average them for an average effect.



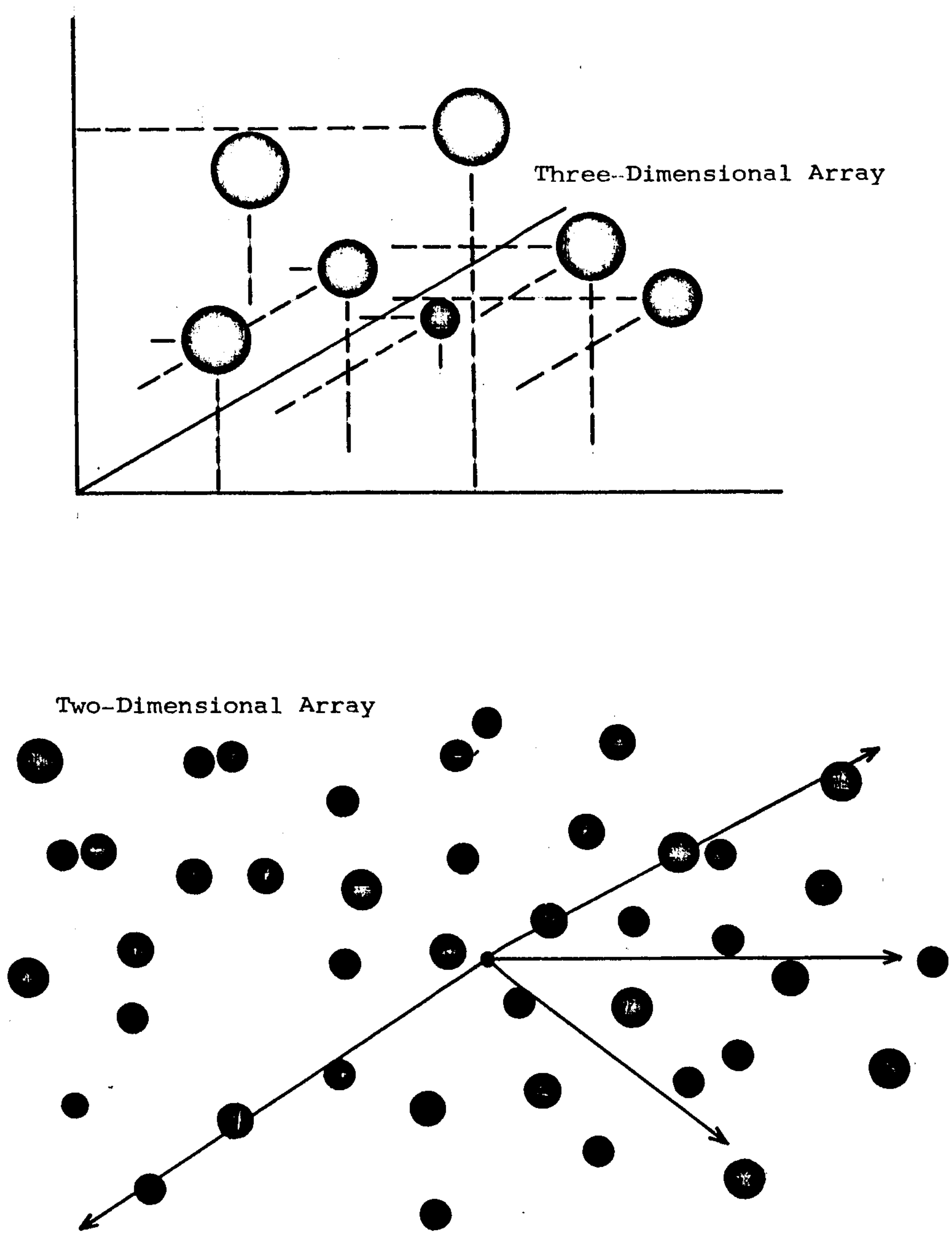
Three-dimensional Section of the Pigment-matrix Array



Two Dimensional Representation of Energy Penetration of the Array

Figure 1

IDEALIZED ARRAY OF MONOSIZED SPHERICAL PARTICLES



A random array has properties independent of the direction studied if averaged over sufficient distance; i.e. a line drawn in any direction will on average intercept the same number of particles.

Figure 2

RANDOM ARRAYS

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Consider radiation incident on a particle. After interaction the energy will be radially distributed about the center of the particle. Then consider possible locations of the next sphere encountered and determine how the energy is distributed. For instance, in Figure 3 the center of the prime scattering is denoted by O. The arrow AO denotes the direction and magnitude of the incident radiation. The other arrows, OB, OC, OD, etc., denote the magnitude and direction of the scattered radiation in the various angular bands delineated by the dotted lines. The distance to the next sphere encountered in any direction will vary depending on the solids concentration of the array and on random fluctuations in position. Since all directions have the same average properties, movement in the first instance can be considered to be in the normal direction.

Let G be the center of the second scattering sphere when the scattering spheres touch and H the center at the greatest probable distance. For any given concentration of solids it should be possible to calculate the probability distribution of the second scattering centers between G and H. For each position of the scattering center along GH the solid angle substituted by the second sphere can be calculated, and the energy in this case can be considered to interact with the second sphere. By averaging for all possible locations and by weighting the average to allow for the probability of occurrence, the average energy occurring at I (the edge of the sphere of influence of the second interaction) can be calculated. Since all directions

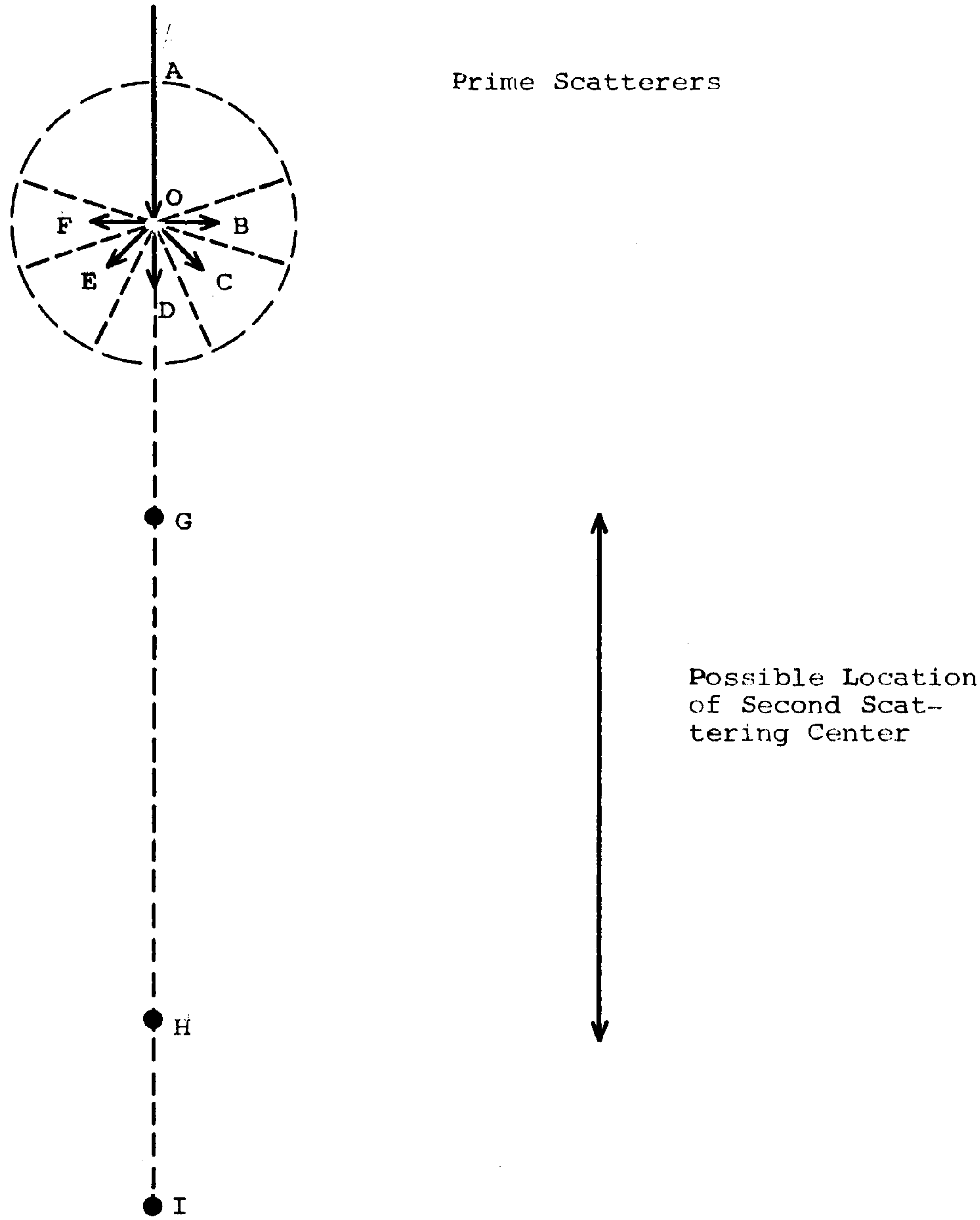


Figure 2

SCHEMATIC DIAGRAM OF INCIDENT RADIATION ON A PARTICLE of incident energy are equally probably and since all second directions are equally probably, the energy at I could represent the average decay in energy for two interactions.

Several aspects of this model need to be examined in great detail, but at this stage it seems to be a reasonable model with which to tackle the scattering problems of a multiple array of particles. In theory it seems possible that the model could be used to study a pigment with a range of sizes. It could also be used to study short-range or long-range penetration.

III. EXPERIMENTAL STUDIES

A. Introduction

The immediate objective of the experimental studies was the establishment of quantitative relationships between particle concentration, the size distribution, and optical properties of light-scattering films. The experiments were limited to quantitative transmittance measurements for various particle-suspension media configurations. These measurements were made on the Cary model 14 recording spectrophotometer. Quantitative measurements of reflectance and of radial intensity distribution of scattered light in highly controlled particle arrays will be given in future reports. In general, two types of suspensions were used for the measurements: (1) dilute suspensions, which simulated idealized scattering systems and (2) thin layers of concentrated suspensions in gelatin, which simulate real pigmented paint systems.

B. Preparation of Monodisperse Sols

The majority of problems with the preparation and size control of uniform silver halide particles were solved during the last research period. Batches of silver bromide with particle diameters varying from 0.1 to 1.0 μ were prepared. The double infusion apparatus and precipitation methods used described in previous reports.[1] The preparation conditions are summarized in Table 1, and Figure 4 shows electron micrographs of the particles. Optimum stability and size control were obtained with silver bromide crystals; they did not grow significantly during several weeks of storage in the refrigerator.

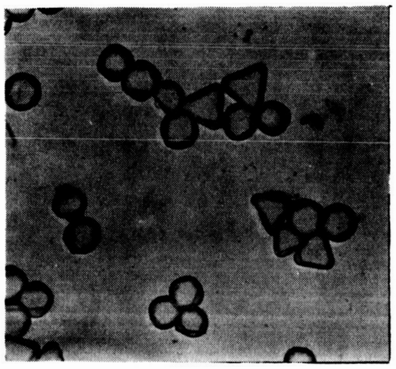
Table 1

PREPARATION OF SILVER BROMIDE SUSPENSIONS

Batch No.	Diam., μ	Normality of KCl and $AgNO_3$	Addition Rate, ml/min	Reaction Conditions Temp., °C	Reaction Conditions pH	Excess KCl ml
25	0.78	2	0.8	58.0	9.6 (NH_3)	2
26	0.64	2	2.0	50.0	9.7 (NH_3)	2
27	0.92	2	2.0	55.0	9.9 (NH_3)	2
28	0.88	2	2.0	55.0	9.7 (NH_3)	2
29	0.92	2	2.0	55.0	9.9 (NH_3)	2
30	0.40	2	2.0	50.0	9.5 (NH_3)	2
31	0.68	2	2.0	50.0	9.7 (NH_3)	2

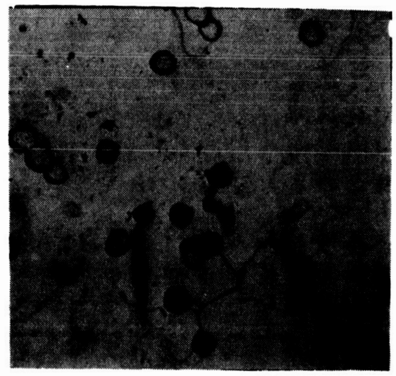
Several batches of monodisperse polystyrene spheres were obtained from the Dow Chemical Company. The electron micrographs

1 Zerlaut, G. A., "Investigations of Light Scattering in Highly Reflecting Pigmented Coatings," Quarterly Report No. IITRI-C6018-6, p. 12, IIT Research Institute, Chicago, Ill., January 1964.



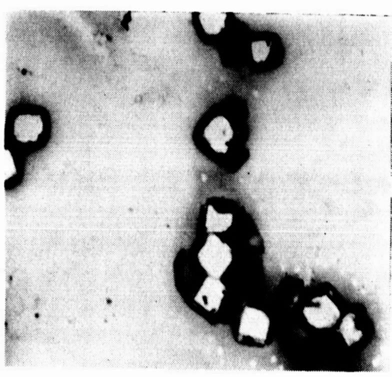
Batch 25

0.78 Micron Diameter
(5000X)



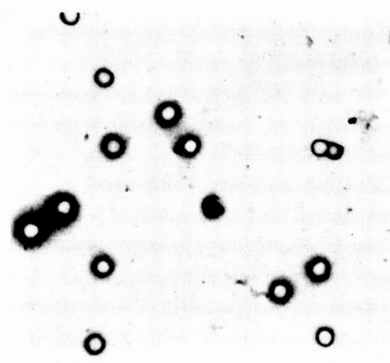
Batch 26

0.64 Micron Diameter
(5000X)



Batch 29

0.92 Micron Diameter
(5000X)



Batch 30

0.40 Micron Diameter
(5000X)

Figure 4

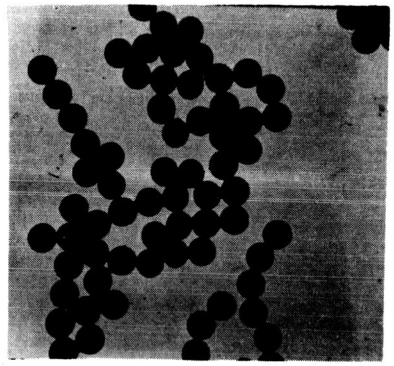
ELECTRON MICROGRAPHS OF SILVER BROMIDE PARTICLES of two particle sizes of polystyrene spheres are shown in Figure 5. The diameters reported by Dow were 1.305 and 0.796 μ with standard deviations 0.0158 are 0.0083 μ, respectively. The polystyrene spheres are of extremely uniform diameter and regular shape. Only the two largest particle sizes could be used, since the smaller particles scatter light at wavelength ranges which are too short for present measurement techniques.

C. Measurements of Optical Properties

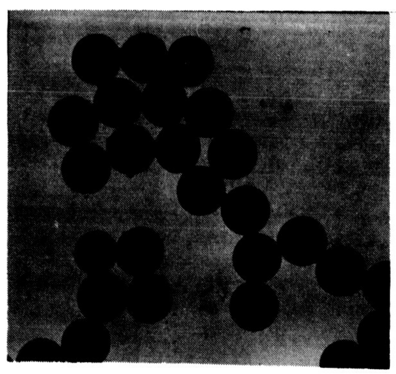
Before studying particle interactions due to concentration effects and size variation e.g., polydisperse systems, it is necessary to know how closely the actual laboratory measurements will approximate the predictions of the Mie theory. It may be useful at this point to reconsider the physical assumptions inplicit in the Mie theory and the relationship of predicted physical quantities to experimental measurements.

The scattering of light is defined as a change in the direction of the photon after interaction with the scattering particle. An inherent assumption significant in experimental measurements is that the theory is valid for infinitely dilute suspenions of monodisperse spheres illuminated monochromatically.

The intensity of a parallel beam of light is reduced in traversing a dispersion of uniform spherical particles according



0.796 Micron Diameter



1.305 Micron Diameter

Figure 5

ELECTRON MICROGRAPHS OF MONODISPERSE POLYSTYRENE SPHERES to the transmission equation in terms of optical density.

$$I/I_o = e^{-K\pi r^2 n l} \tag{1}$$

or

$$D = K\pi r^2 n l \tag{2}$$

The form of Equation 1 is very similar to the Lambert-Beer law of absorption:

$$I/I_o = e^{-a n l} \tag{3}$$

where $K\pi r^2$ is replaced by a. Here K is the Mie scattering coefficient, and n is the particle concentration term in Equation 1 and a general concentration term in Equation 3.

The spectral transmittance curves, predicted by Mie theory, are therefore oscillatory with the minima and maxima of transmittance which correspond inversely to the oscillations in K. Since K is a function of size parameter α, where $\alpha = 2\pi r/\lambda$ for monodisperse particles of different radii, the minima and maxima will occur at different wavelengths.

Thus, with the use of spectral transmittance measurements and Equation 1 the radii of the particles can be determined from the wavelength positions of the minima and maxima, and the number (concentration) of scatterers can be determined from absolute transmittance at any given wavelength. The reciprocity of path length concentration is assumed in Equation 1.

D. Particle Size Measurements from Light Scattering

Figure 6 shows the spectral transmittance curves for the majority of batches of silver bromide particles used in this study. These curves were obtained from water suspensions containing approximately equal weights (o.707 g/ml of silver bromide particles by using a 2-cm path length absorption cell.

The first transmittance minimum (measured from the long-wavelength side) corresponds to the first scattering maximum; the first transmittance maximum crorresponds to the first scattering minimum; etc.

Figure 7 summarizes graphical particle size measurements from light scattering and indicates wavelength positions of extrema derived from the theory for refractive index of 1.7. It is the effective refractive index of silver bromide in water, calculated from

$$m = \frac{m(\text{particle})}{m\ (\text{medium})} = \frac{2.253}{1.333} = 1.7$$

A consistent increase in measured radii at higher lobes can be seen in Figure 7. This deviation could be due to the following effects: (1) the absorption coefficient of silver bromide increases significantly at shorter wavelengths, and the effective refractive index becomes a complex number which is not considered in our assumption of constant refractive index (1.7). (2) it can be shown that the deviations due to nonuniformity of the particles would be significant at higher lobes.

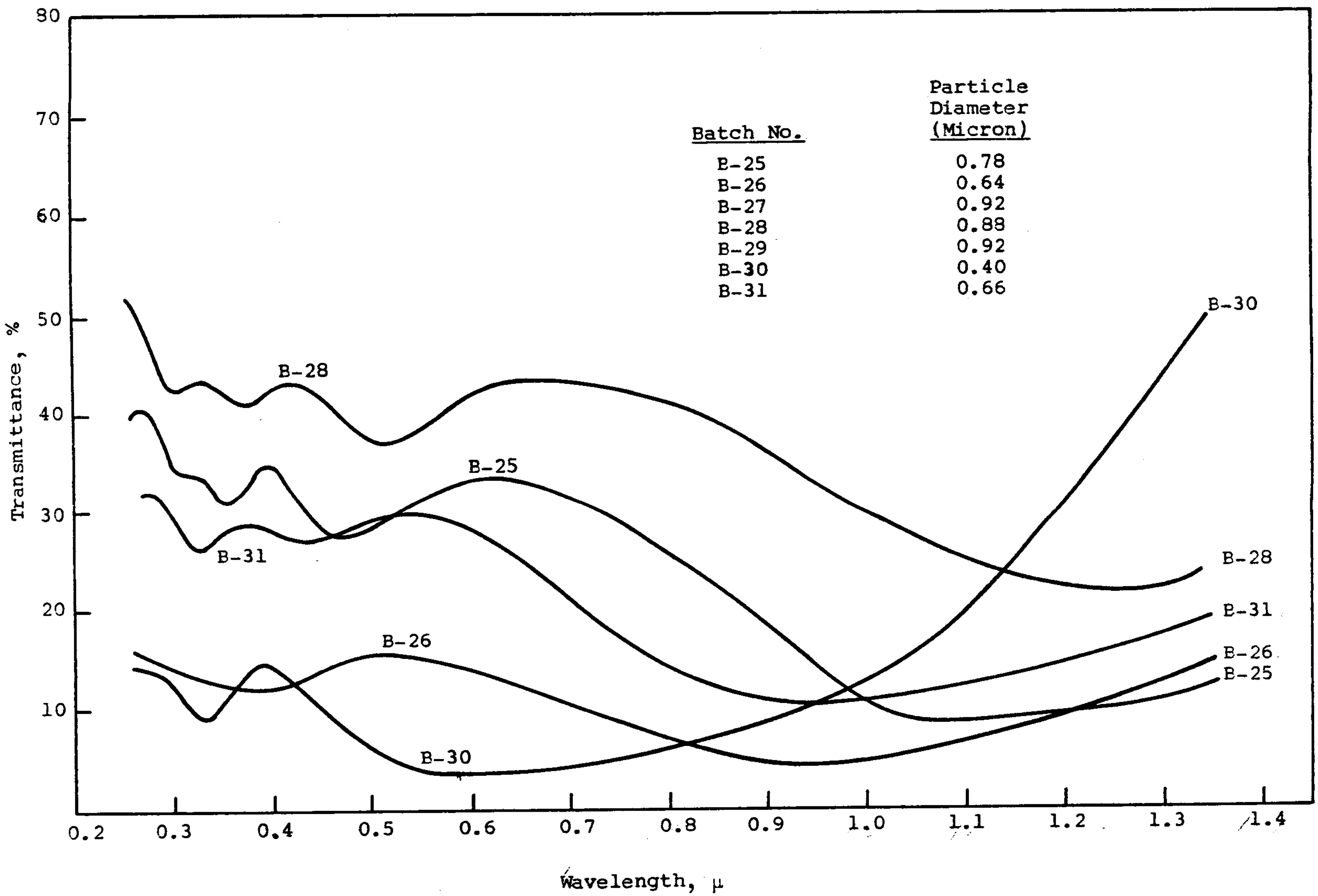


Figure 6

TRANSMITTANCE OF SILVER BROMIDE SUSPENSION IN WATER

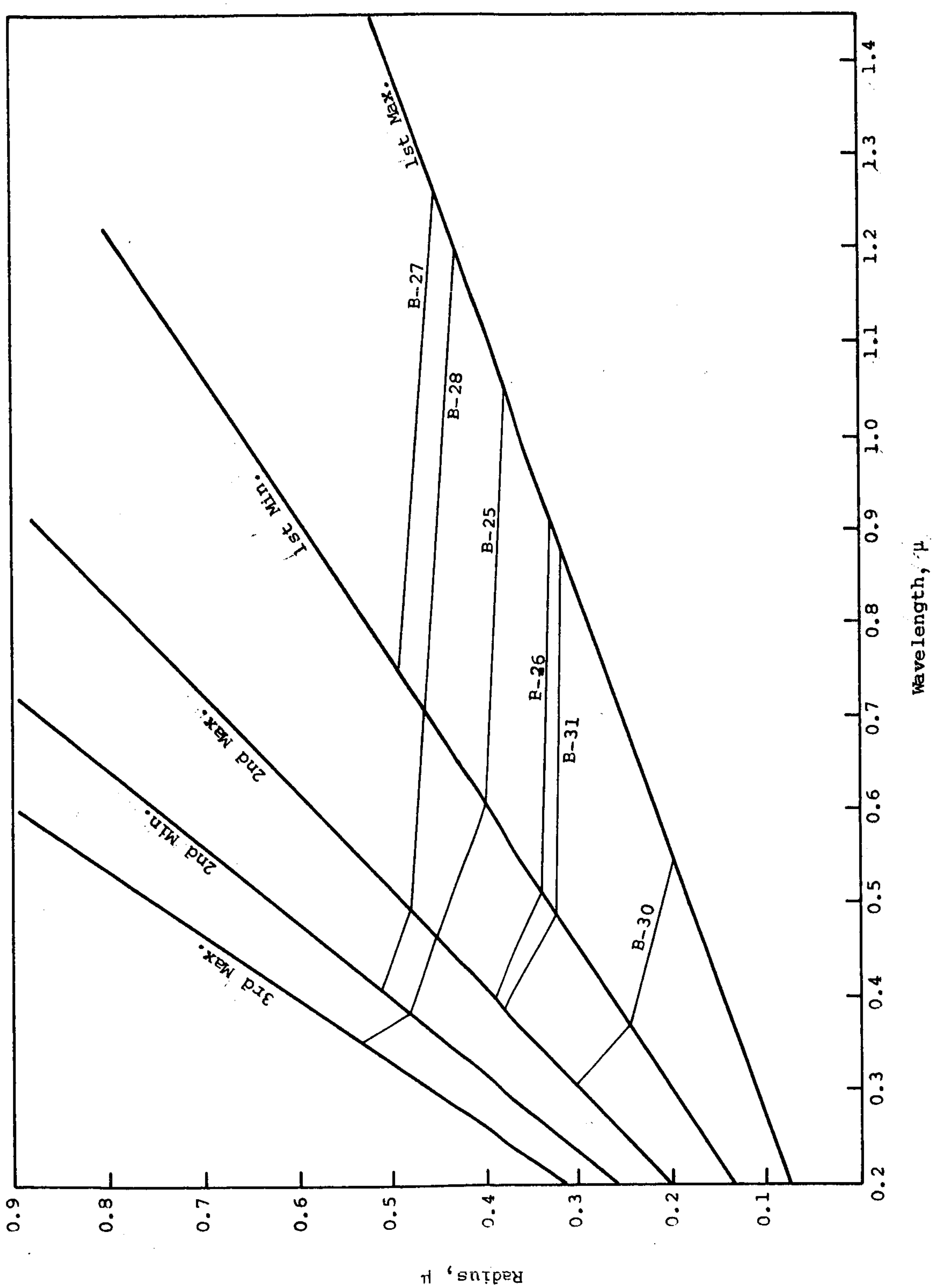


Figure 7

PARTICLE SIZE MEASUREMENTS FROM LIGHT SCATTERING

Figures 8 to 12 shows some actual count distributions from electron micrographs of the particles which are compared with the geometric mean and the light-scattering measurements.

Table 2 summarizes the data from the particle size measurements. Particle radii were measured from (1) light scattering (graphically), (2) light scattering calculated numerically from the normalized size parameter $\rho = 2\alpha(m-1)$, and (3) electron micrograph geometric mean.

Table 2

PARTICLE RADII MEASUREMENTS

	Radius of Particles			
	Measured by Light Scattering			Measured by Electron Micrograph,
	Numerically			
Batch No.	First Max.	First Min.	Graphically	Geo. Mean
25 AgBr	0.384	0.407	0.39	0.32
26 AgBr	0.319	0.316	0.32	0.28
27 AgBr	0.456	0.513	0.46	0.49
28 AgBr	0.431	0.421	0.44	0.38
29 AgBr	0.456	0.513	0.46	0.49
30 AgBr	0.194	0.250	0.20	0.20
31 AgBr	0.337	0.349	0.33	0.34
Polystyrene	0.427	0.502	0.43	0.398
Polystyrene	0.686	0.742	0.72	0.653

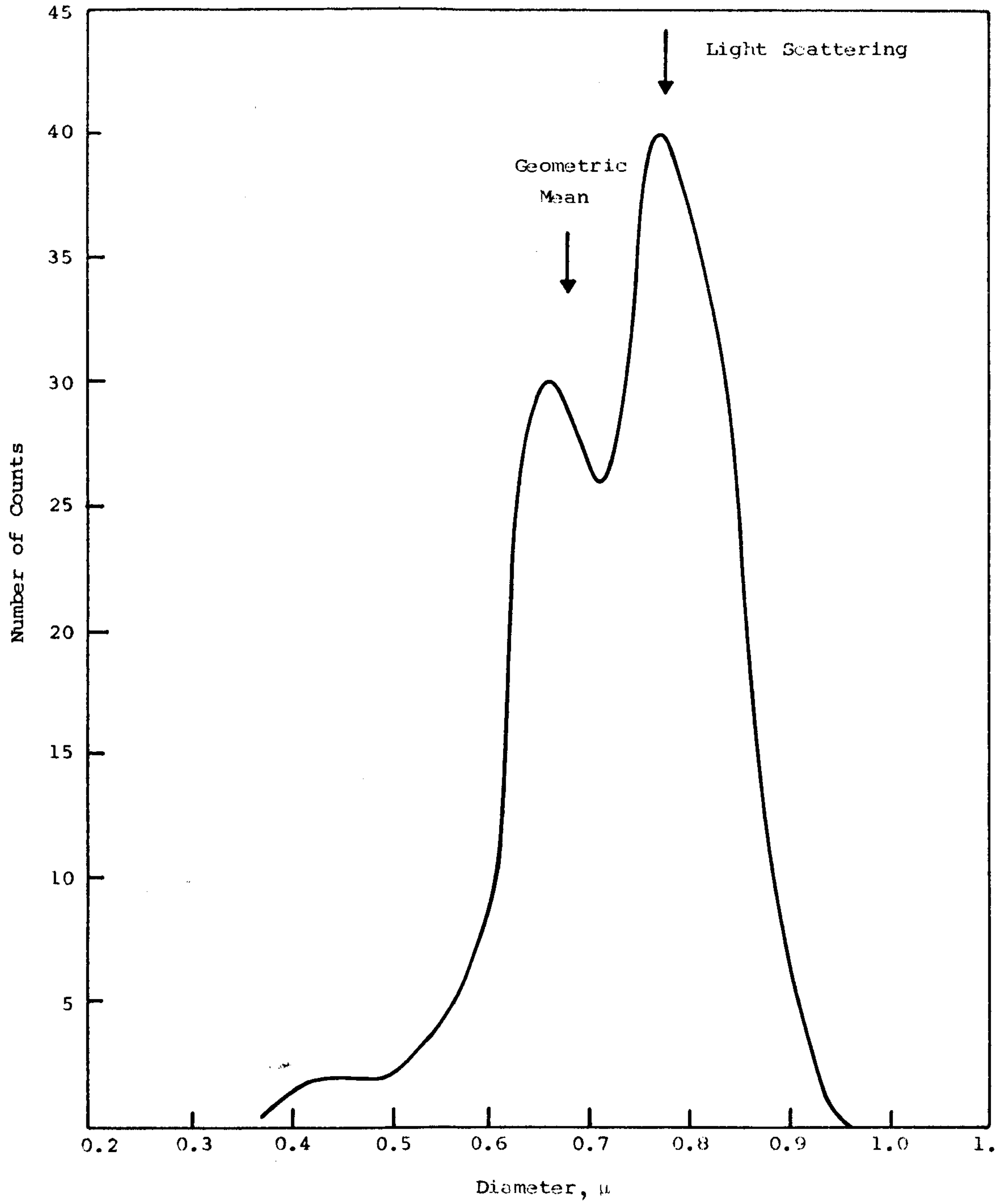


Figure 8
PARTICLE SIZE DISTRIBUTION FOR BATCH 25

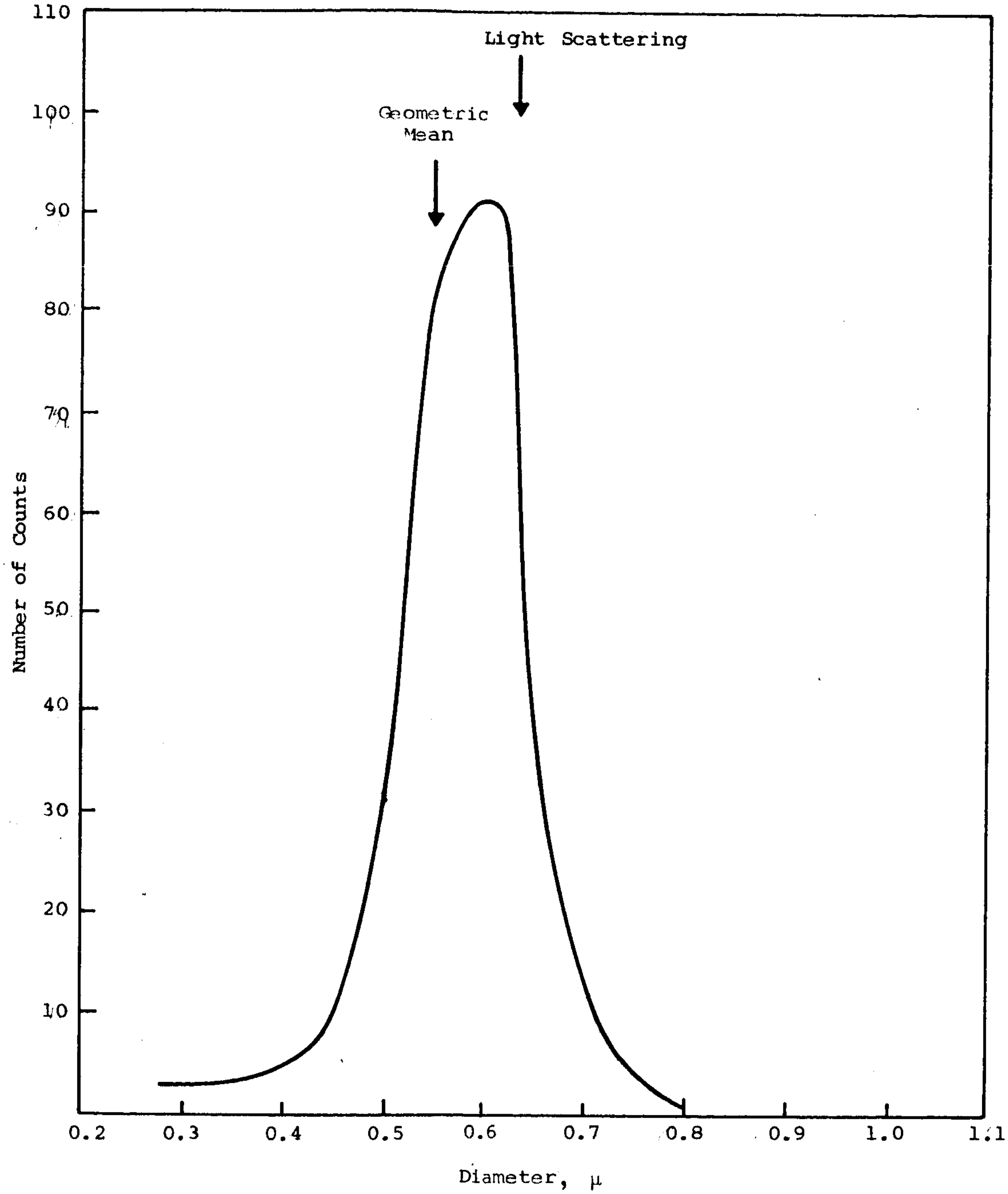


Figure 9
PARTICLE SIZE DISTRIBUTION FOR BATCH 26

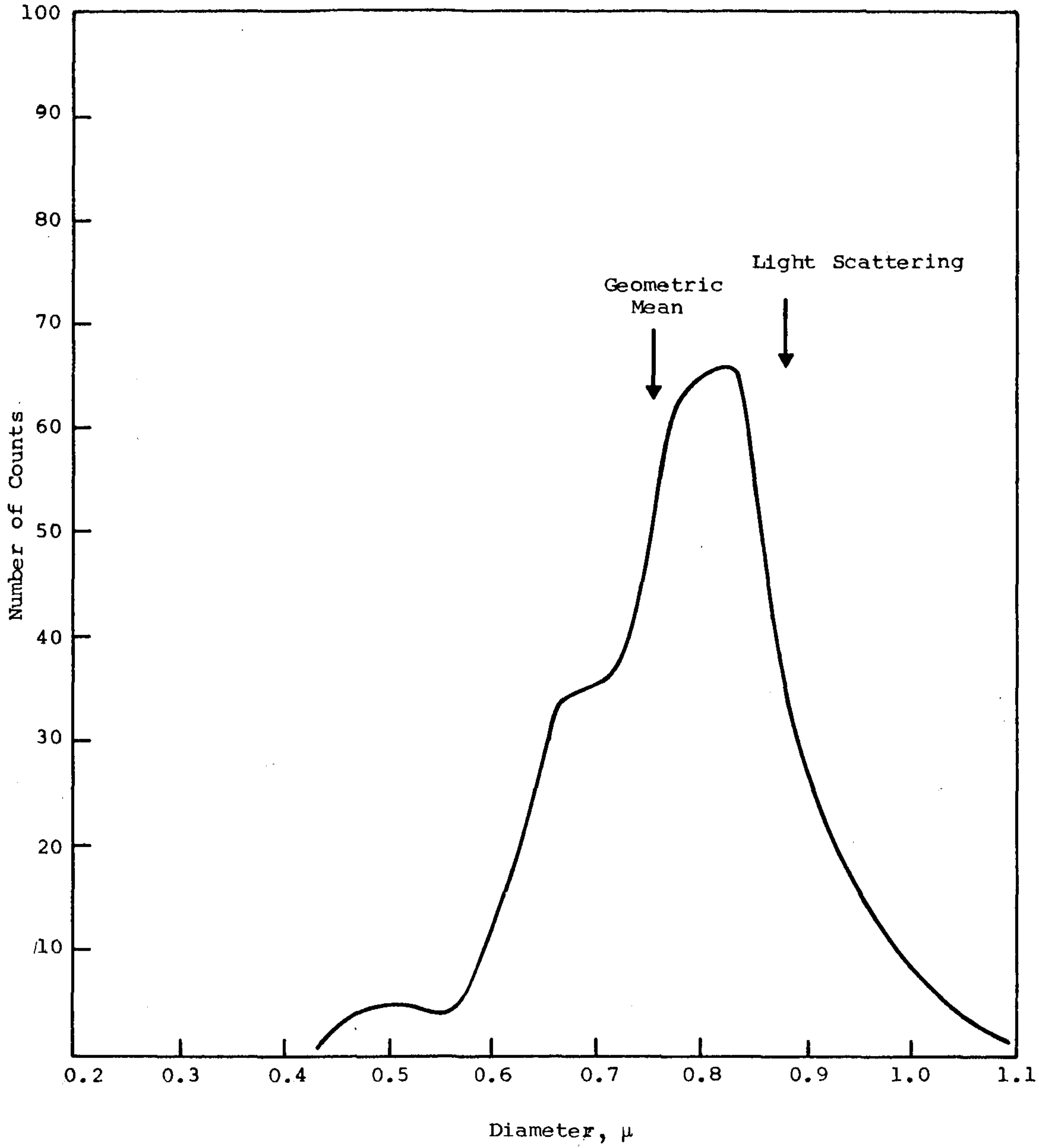


Figure 10

PARTICLE SIZE DISTRIBUTION FOR BATCH 28

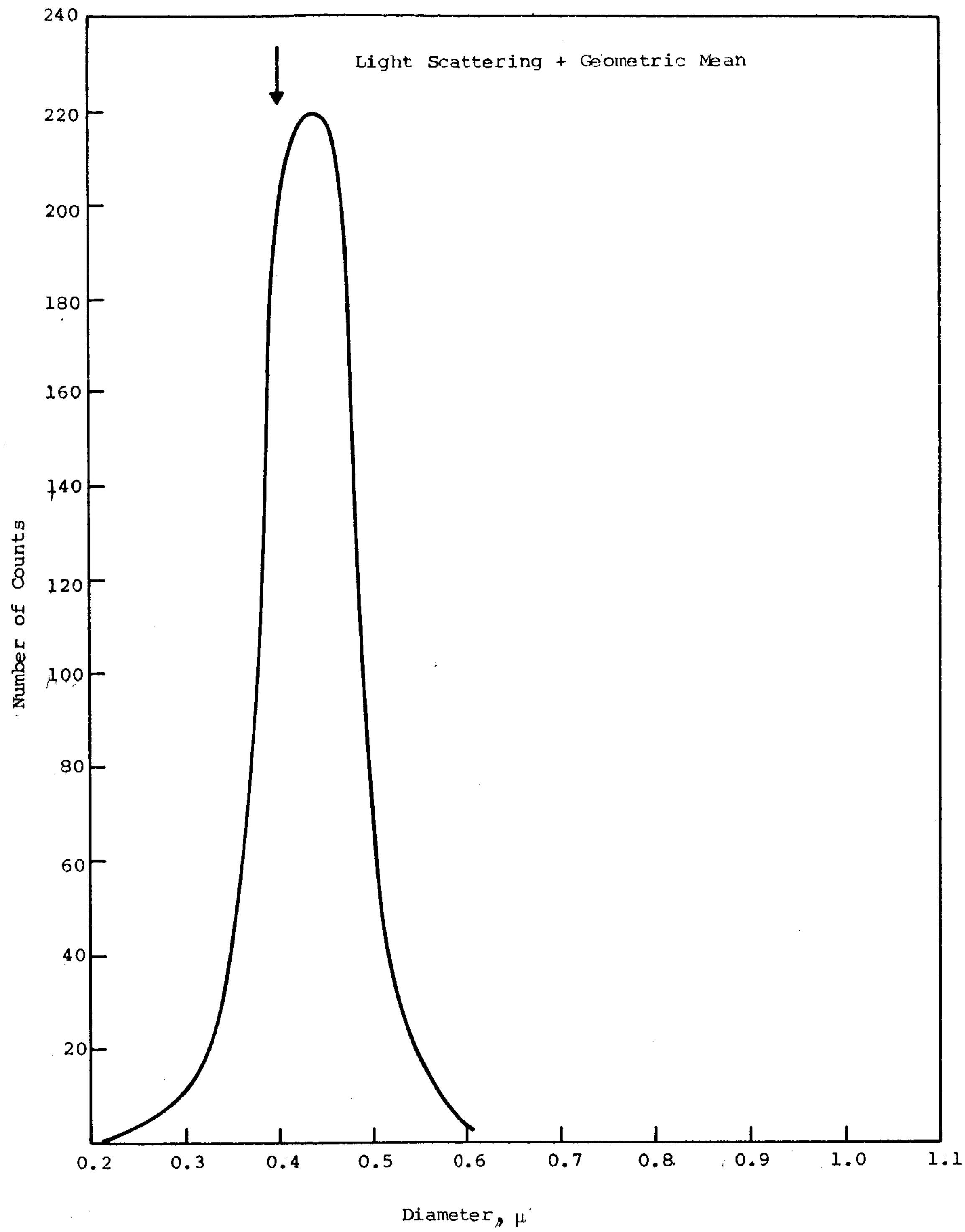


Figure 11

PARTICLE SIZE DISTRIBUTION FOR BATCH 30

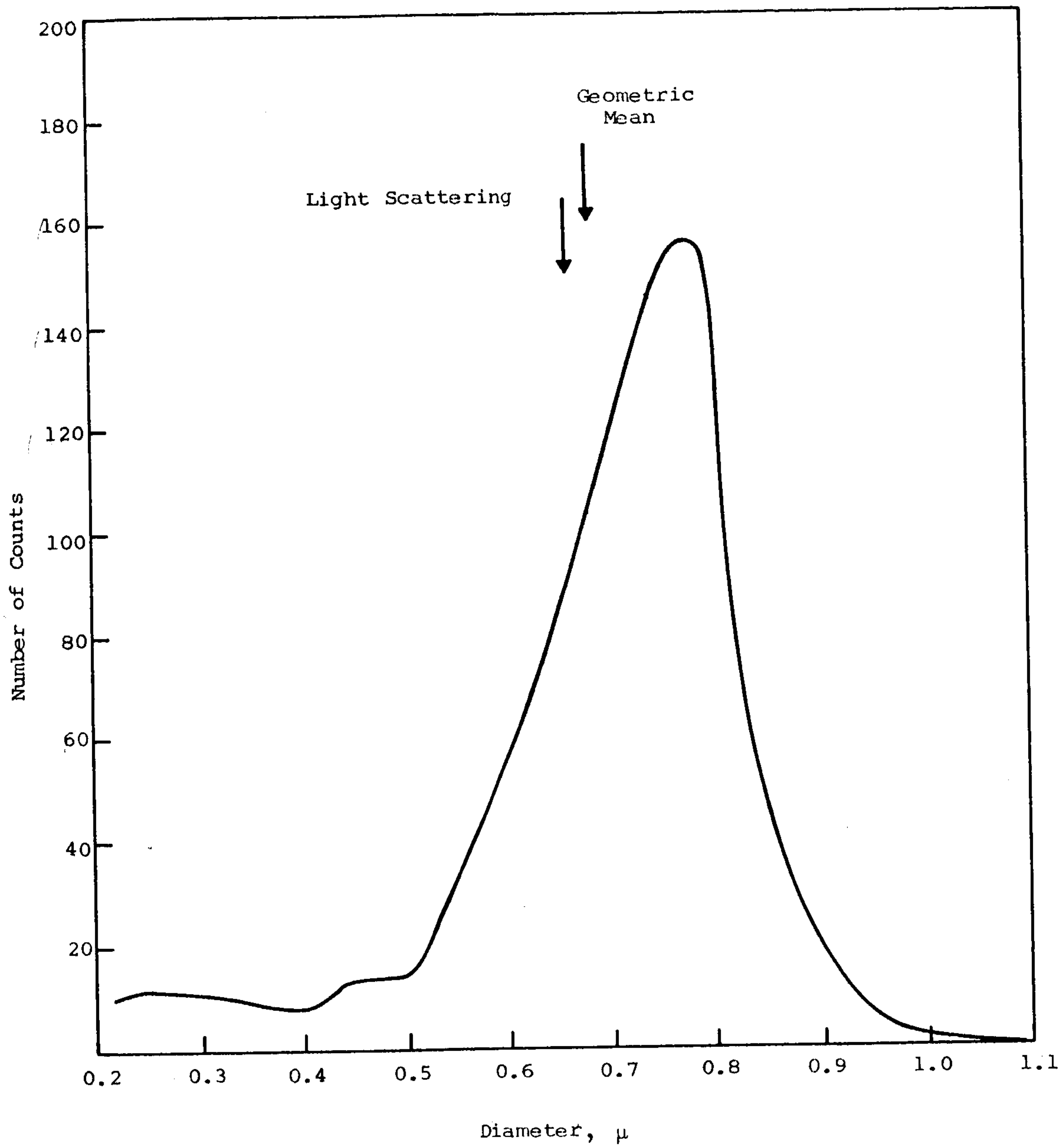


Figure 12

PARTICLE SIZE DISTRIBUTION FOR BATCH 31

E. Concentration Measurements of Dilute Suspensions

After the radii of the particles have been determined, the number of scatterers per unit volume can be determined from the transmission equation, (Equation 1) and from transmittance measurements at any given wavelength. The concentration of silver bromide particles in dilute suspension were measured from the transmittances at several wavelengths and at varying dilutions. The wavelengths were chosen to correspond to the first scattering maximum, the first mimimum, and the midpoint between them. Since the size parameter, α, is a reciprocal function of wavelength, λ, the midpoint corresponds to the midpoint of the wavelength reciprocals of maximum and minimum transmittances.

The measured concentrations were compared with concentration independently calculated from the weight of silver bromide or polystyrene in suspension. If the weights and densities of materials in suspension are known, the total volume in suspension is the ratio of weight to density. If the radius (volume) of each particle is known, the number of particles equals the ratio of the total volume of material to the volume of one particle. It should be emphasized that in this calculation all particles are assumed to be uniform, perfect spheres; this assumption does not directly apply to silver bromide particles. The data are summarized in Table 3 for silver bromide and polystyrene. Maximum deviations in concentration measurements would be expected at the wavelengths corresponding to the extrema in the total Mie scattering coefficient, K.

Table 3

CONCENTRATION MEASUREMENTS IN WATER SUSPENSIONS

	Concentration, $N^* \times 10^{-8}$			
	Measured by Light Scattering			
Batch No.	First Max.	First Min.	Midpoint	Calculated
25 AgBr	0.586	0.645	0.594	0.586
26 AgBr	1.105	1.56	1.40	1.165
27 AgBr	0.248	0.342	0.271	0.358
28 AgBr	0.402	0.458	0.405	0.408
29 AgBr	0.248	0.342	0.271	0.358
30 AgBr	3.43	4.10	3.72	3.70
31 AgBr	0.75	0.95	0.84	0.87
Polystyrene (0.796)			2.17	2.239
Polystyrene (1.305)			0.522	0.531

* n = no. of particles/cm^3.

Examination of the transmission equation (Equation 2) reveals that the optical density is a linear function of particle concentration for dilute suspensions. Figure 13 shows the observed optical density (to the base of the natural log) versus the independently calculated particle concentration and indicates that the transmission equation is valid up to a particle separation of 17 diameters ($n = 3.06 \times 10^8$ particles/cm^3).

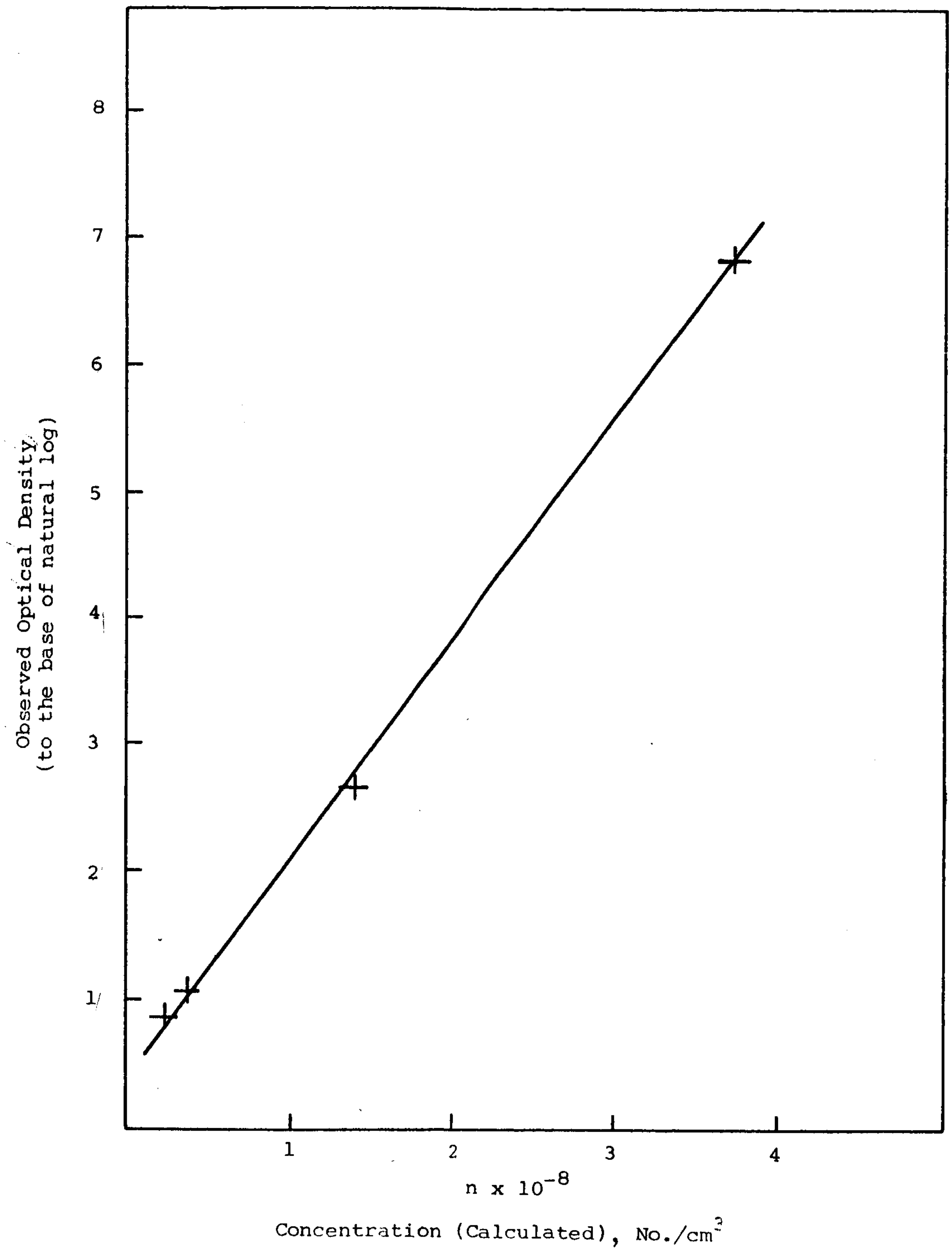


Figure 13
CALCULATED CONCENTRATION vs. OBSERVED OPTICAL DENSITY

An incidental observation may be noted: this separation of particles (1.5 diamters) is approximately the limit for the measurement of transmittance by using 1-cm path length cells. At higher concentrations the transmittance is too low to obtain a reasonable signal:noise ratio.

F. Arrangement of Particles in Gelatin

To attain more closely packed suspensions, approximating real pigmented paint systems, various concentrations of particles were dispersed in gelatin and deposited in thin layers on quartz plates. The thickness of the films was varied; excess suspension was allowed to drain by holding the quartz plates at varying angles. The films were rapidly dried in a hot air stream. A detailed description of the methods of preparation of gelatin films was given in earlier reports.[2]

Since the total Mie scattering coefficient, K, depends on the refractive index of the scattering array, e.g., the ratio of refractive indexes of particles to suspension medium, it is possible to predict the wavelength shifts of the observed minima and maxima of scattering with variation of the refractive index of suspension medium. The size and refractive index of the particles themselves are constant.

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[2] Zerlaut, G. A. et al "Investigation of Light Scattering in Highly Reflecting Pigmented Coatings," Quarterly Report No. IITRI-C6018-6, p. 22, IIT Research Institute, Chicago, Ill. January, 1964.

Therefore, as the suspensions for any given particle size of silver bromide are prepared (e.g., in water, gelatin or ethanol), predicatable wavelength shifts should be observed. For three-dimensional arrays in liquids (dilute suspensions in absorption cells) with different refractive indexes, the observed wavelength shifts gave excellent agreement with these predicted by theory (Figure 6). However, dry gelatin films apparently gave completely erratic wavelength shifts of scattering extrema (in magnitude but not direction). This unpredictable behavior in thin layers in gelatin was subsequently explained as a result of thickness measurements which correspond to path length in an absorption cell. The thicknesses of the gelatin suspension layers were measured interferometrically by using a Zeiss model 2300 interference microscope. A clean scratch was made in the gelatin layer, and in a majority of cases three interference patterns could be detected:

1. due to the quartz substrate
2. due to the gelatin layer
3. a very faint one, due to the particles themselves.

The most significant quantitative observations from the thickness measurements were: (1) the particles were imbedded in the layers of gelatin, which were considerably thinner than the particles themselves, and (2) in a majority of cases the particles were arranged (packed) in monolayers. The manner of packing is schematically represented in Figure 14.

Since the layers are very thin (0.2 to 2.0 μ), the accuracy of the thickness measured is within ± 15%. Also, because the particles are not completely surrounded by gelatin, the effective refractive index varies with the thickness of the gelatin layer.

G. Transmittance of Monodisperse, Concentrated Films

In visualizing the transmittance characteristics of monodisperse closely packed suspensions of gradually increasing thickness, it is necessary to reconsider the physical mechanism of energy loss. It would be unreasonable to expect that the transmittance losses of parallel light in thick layers of concentrated suspensions would be similar to those occurring in dilute suspensions, e.g., due only to changes in the direction of photons.

From theoretical considerations, it is expected that as the number of closely packed layers increases, the number of scattering events (changes in the direction of incident photons) will also increase. Thus increasing amounts of light in the deeper layers will be lost by absorption and will be converted to thermal and molecular energy modes. An increasing number of scattering events will lead to an increasingly effective absorptive path length for each penetrating photon. Thus even at low absorption efficiency (as is the case of silver bromide), if the absorption path is long enough, the

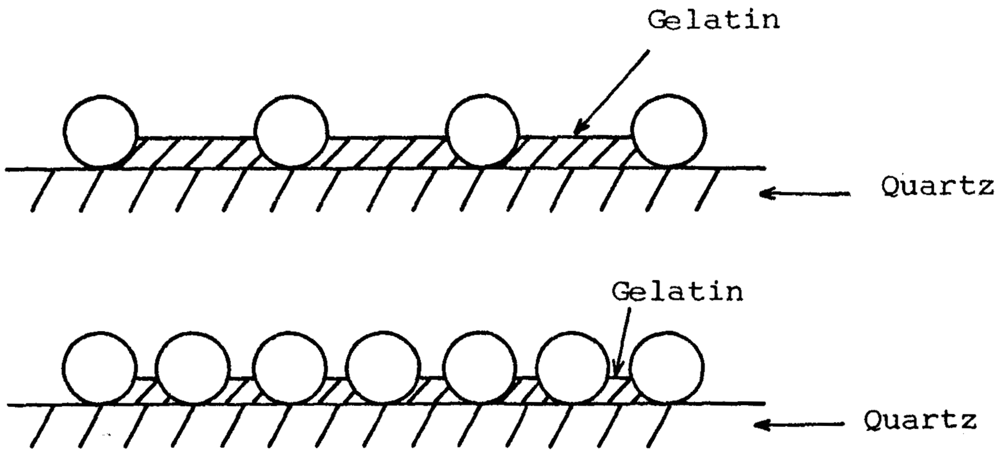

Figure 14

SCHEMATIC REPRESENTATION OF PARTICLE PACKING
IN GELATIN LAYERS AT VARYING CONCENTRATIONS

absorptive mechanism (Beer-Lambert's Law) of energy loss will predominate in the deepest layers of the suspension.

The resultant experimental observation is a gradual decrease in the amplitude of the scattering minima and maxima and finally a transition into a continuous function similar to Lambert's law of absorption. In the longer wavelength region where the scattering is minimal, the effective absorptive path length corresponds to approximately a single pass of the photon. Since there is no appreciable change in direction of the photon due to scattering, the absorptive losses are very slight, and the transmittance increases significantly. Figure 15 shows the source of the experimental spectral transmittance curves of concentrated suspenions as a function of film thickness.

At short wavelengths (Figure 16) the amplitude of extrema and the total trasmittance decrease with increasing thickness. At longer wavelengths a sharp increase may be noted because scattering, and therefore absorption, becomes minimal. This very strong wavelength dependence is also characteristic of Rayleigh scattering; scattered energy is proportional to λ^{-4}.

An actual increase in transmittance at wavelengths corresponding to the first scattering maximum with increasing concentration can be observed in several intermediate thicknesses (Figure 17). This phenomenon is also thought to be caused by multiple scattering events. In the intermediate

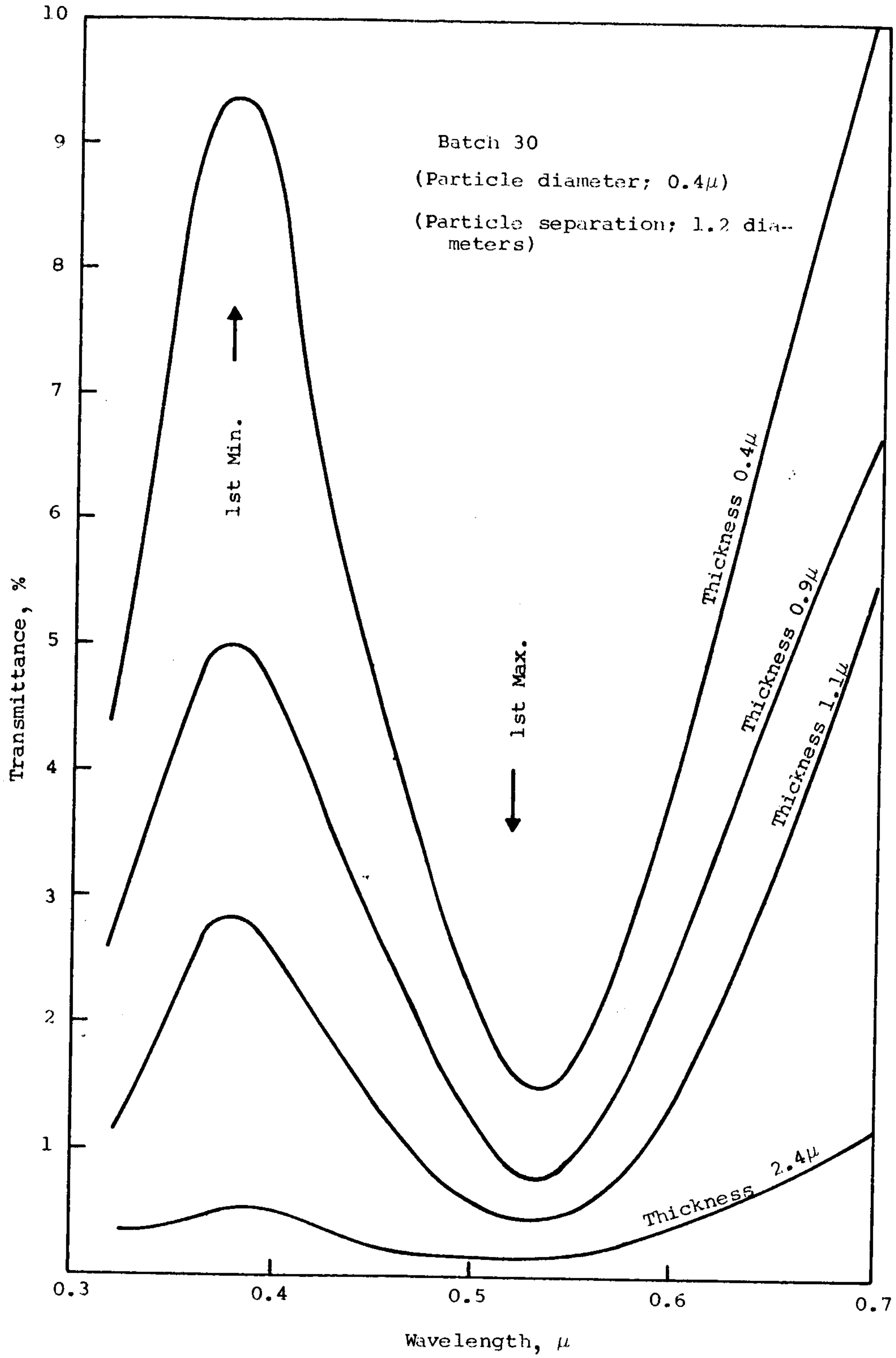

Figure 15

SCATTERING EXTREMA vs. THICKNESS OF GELATIN SUSPENSION

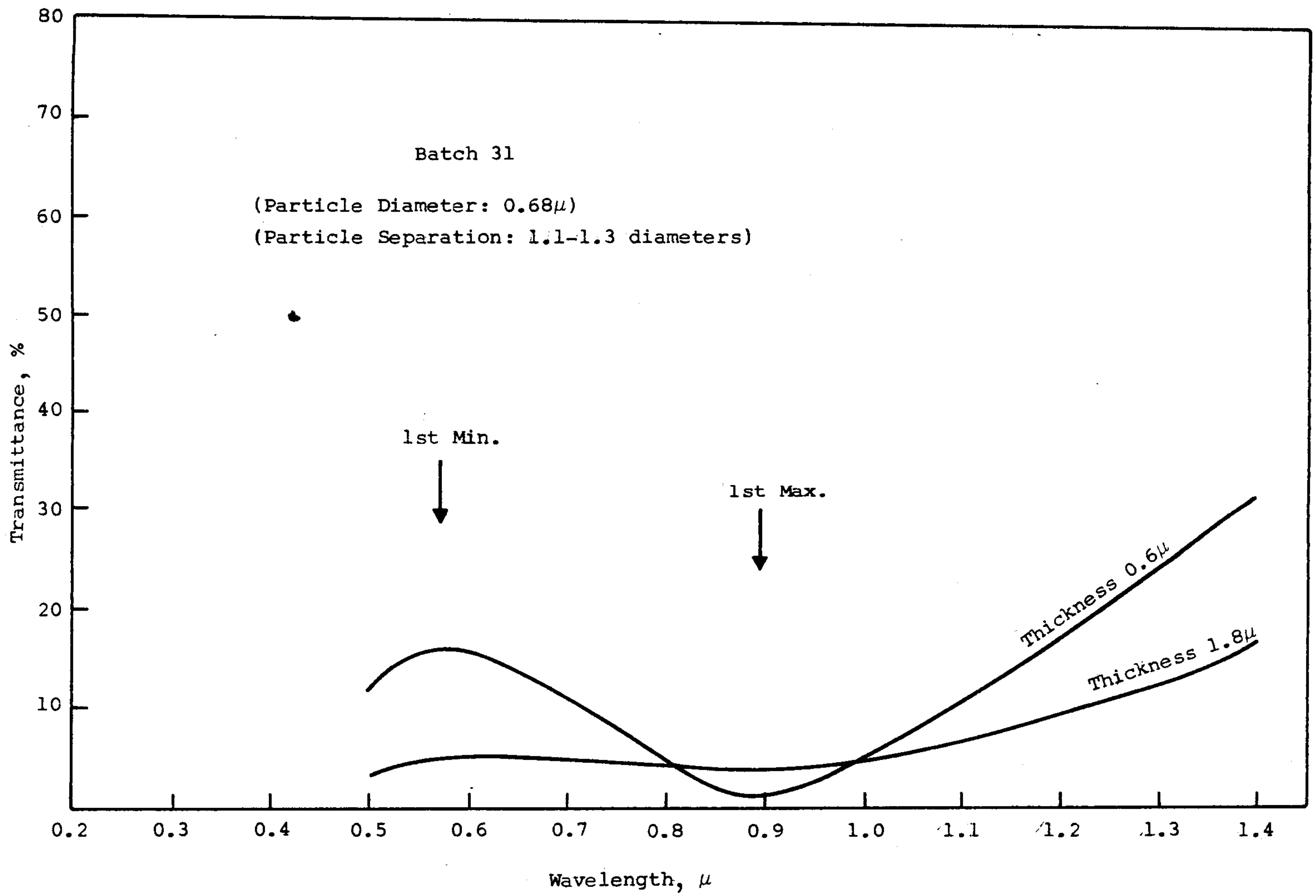

Figure 16

SPECTRAL TRANSMITTANCE OF CONCENTRATED GELATIN FILMS

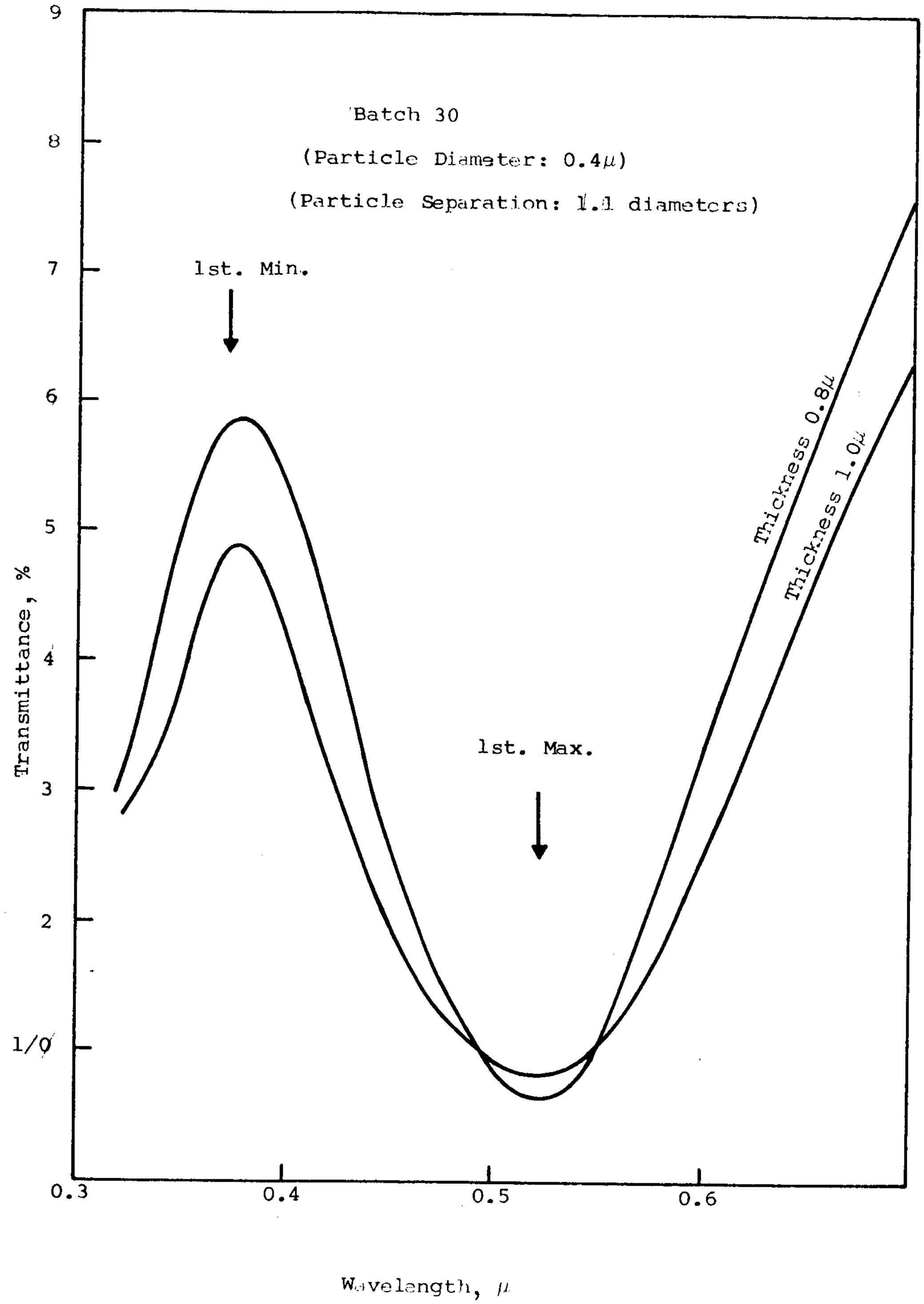

Figure 17

SPECTRAL TRANSMITTANCE OF CONCENTRATED GELATIN FILMS

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cases, the decrease in the amplitude of transmittance minima and maxima is probably the best indication of when the single scattering mechanism begins to fail and multiple scatter and absorption becomes the dominant mechanism of energy loss.

The ratios of extrema for various particle sizes, thicknesses, and concentrations of films are summarized in Table 4. Only very slight interactions occur independently of concentration if the particles are suspended in monolayers, e.g., thin films with particles packed side by side with respect to the incident beam of light. However, in multiple layers (thick films) even within three layers the deviations become quite significant.

The above observation is further confirmed by the measurements of dilute suspensions in three-dimensional arrays with long path lengths. In this case a slight but definite decrease in the ratio of extrema was observed.

The conclusion that even extremely close packing of particles side by side in monolayers gives only very slight interactions can be quantitatively explained by considering the radial intensity distribution functions. In general, the radial intensity functions approximate the figure-eight with an intensity minimum approximately 90° from the direction of the incident beam. Thus minimal interaction would be expected if the particles are packed side by side.

Table 4

OPTICAL DENSITY AND RATIO OF SCATTERING EXTREMA IN MONOLAYER SUSPENSIONS

Relative Dilution: AgBr Suspension/Gelatin	Density First Max.	Density First Min.	Density Second Max.	Ratio First Max./First Min.	Ratio Second Max./First Min.
Batch 31: * 1/2	0.810	0.432	0.615	1.875	1.424
1/1	1.220	0.609	0.935	2.003	1.535
2/1	1.405	0.672	.075	2.091	1.600
Theoretical (Mie Scattering)	-	-	-	2.63	1.82
Batch 25:** 1/2	0.777	0.327	0.564	2.376	1.724
1/1	1.238	0.477	0.775	5.595	1.624
2/1	2.810	1.109	1.930	2.534	1.740
Undiluted	4.60	1.965	3.440	2.341	1.751
Theoretical (Mie Scattering)	-	-	-	2.63	1.82

* Film Thickness, 0.6 to 0.8μ ; diameter, 0.66μ ; particle separation (indiluted), 1.1 to 1.3 diamters.

** Film thickness, 0.8 to 1.2μ ; diameter, 0.78μ ; particle separation (undiluted), 1.1 to 1.3 diamters.

H. Transmittances of Suspensions Containing Two Particle Sizes

To approximate polydisperse systems, a series of mixtures containing two particle sizes were prepared. The transmittance curves of dilute suspensions of these mixtures are given in Figures 18 to 21. The data indicate that in dilute systems the particles at two sizes act as independent scatterers. The data show that independent transmittance curves for each particle size intersected with the transmittance curve of the mixture. Sharp discontinuities in the transmittance of the mixture were observed in several cases at the intersection of the three curves. It can be shown algebraically from the transmission equation (Equation 1) that the transmittance of the mixture will equal the transmittances of two components at this single wavelength, although the optical densities are additive at any wavelength.

An exception may be noted in Figure 18 where the curves do not intersect in the short wavelength range but do intersect in the long wavelength range. This can be explained on the basis of increasing absorption of silver bromide at shorter wavelengths. The transmission equation considers the energy losses due only to scattering and not to absorption. We have shown previously that the observed transmittance curves tend to deviate from the predicted ones at short wavelengths when absorption effects become significant.

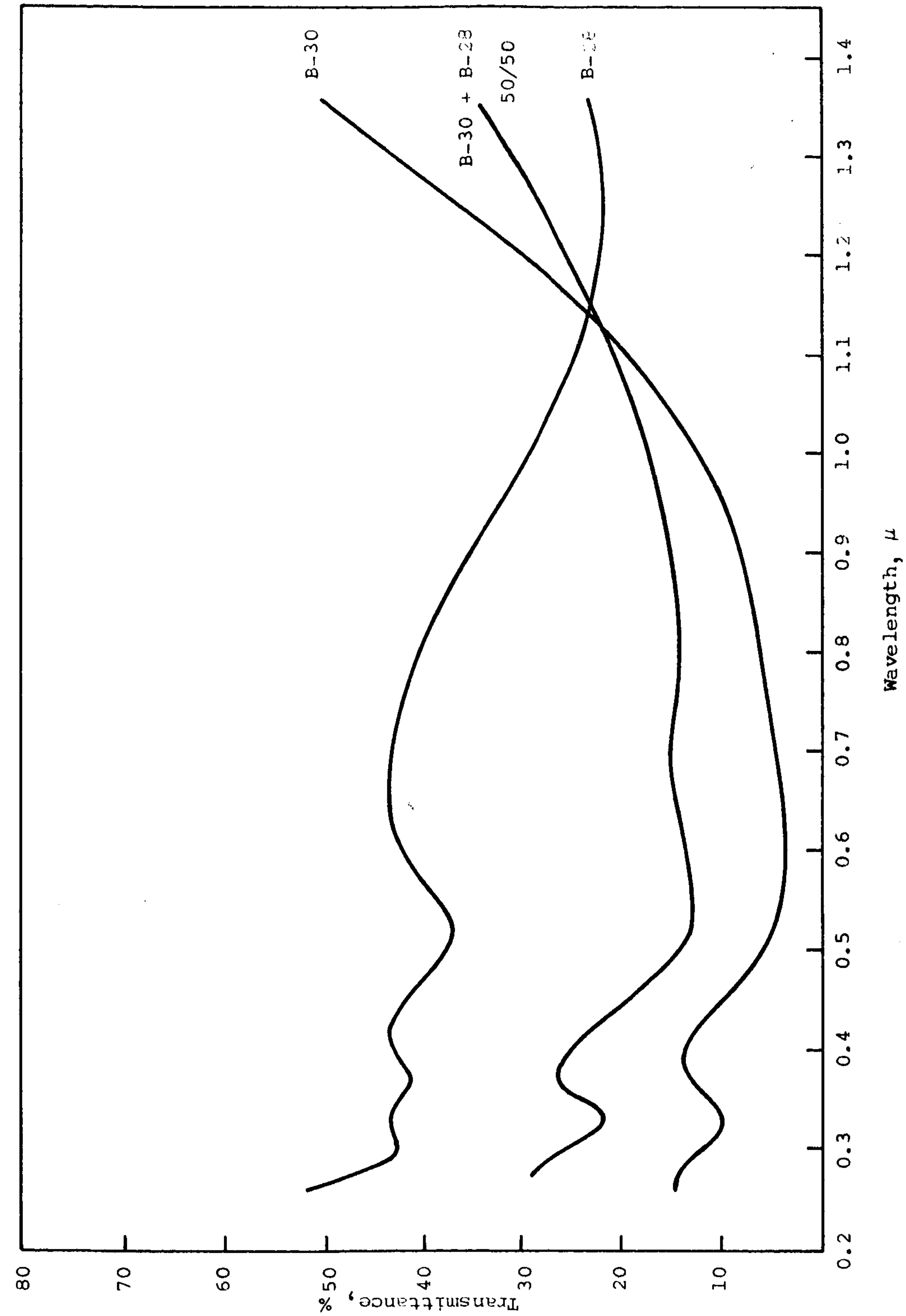

Figure 18
TRANSMITTANCE OF DILUTE SUSPENSIONS OF SILVER BROMIDE MIXTURES
(Batches 28 and 30)

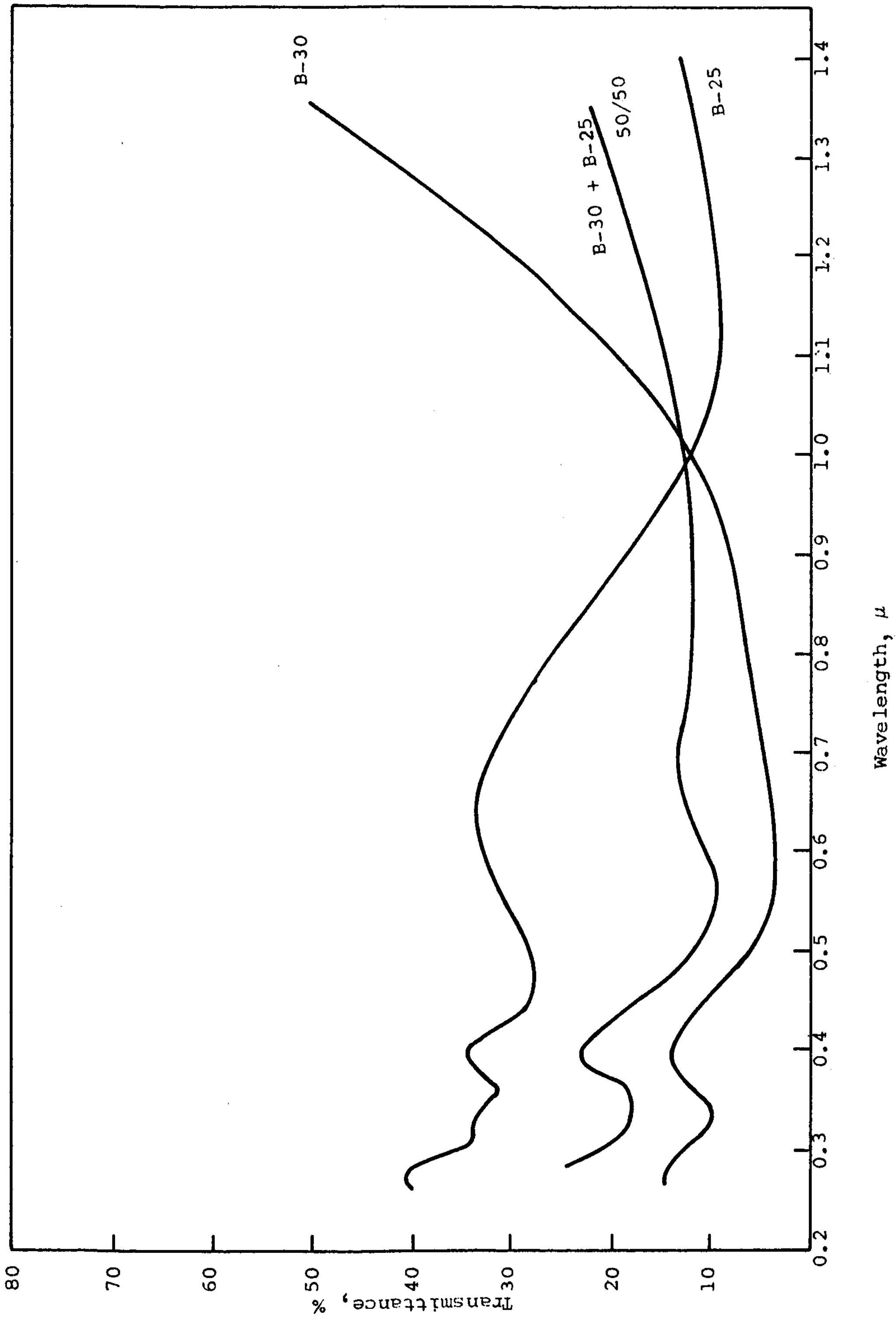

Figure 19

TRANSMITTANCE OF DILUTE SUSPENSIONS OF SILVER BROMIDE MIXTURES
(Batches 25 and 30)

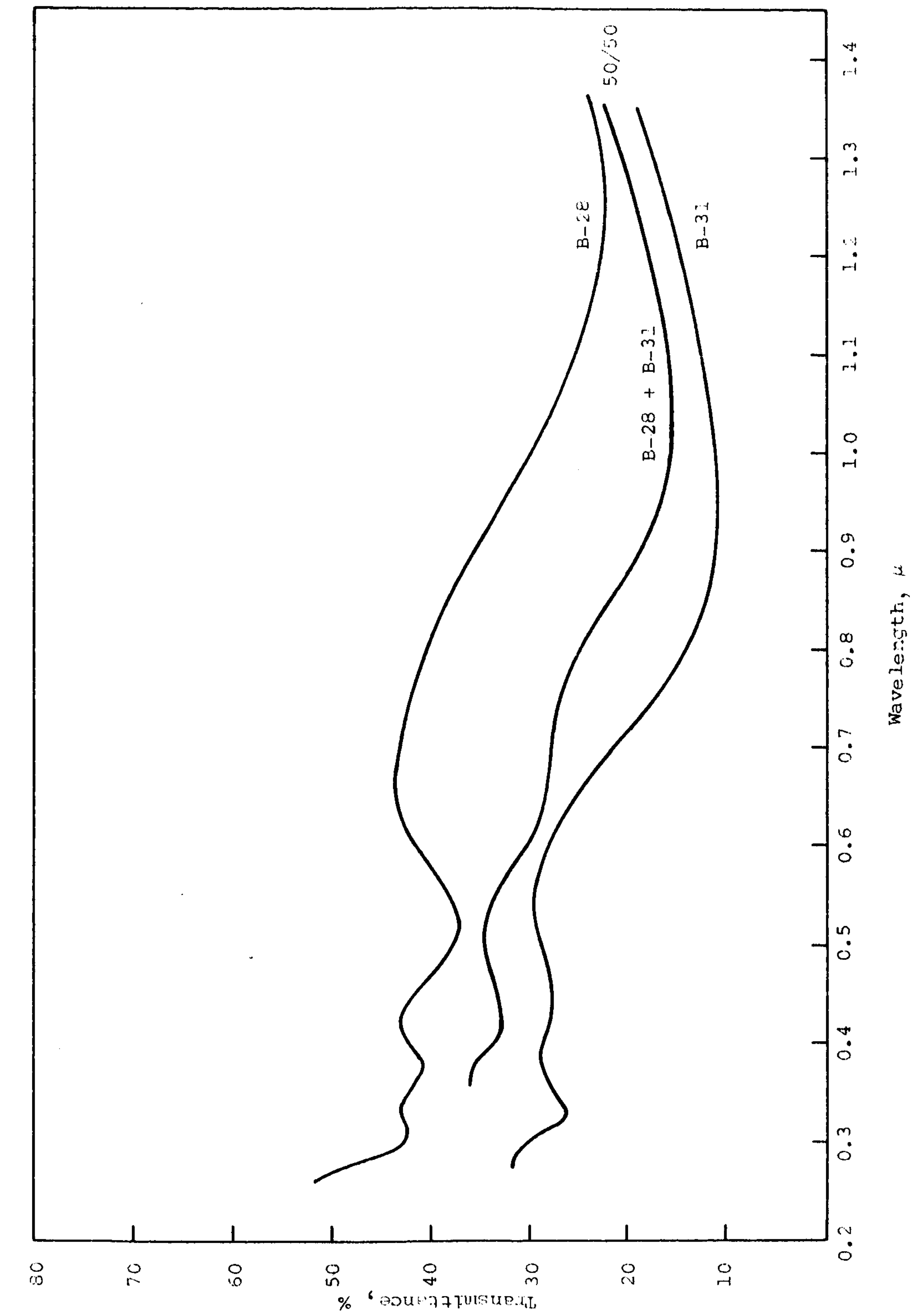

Figure 20
TRANSMITTANCE OF DILUTE SUSPENSIONS OF SILVER BROMIDE MIXTURES
(Batches 28 and 31)

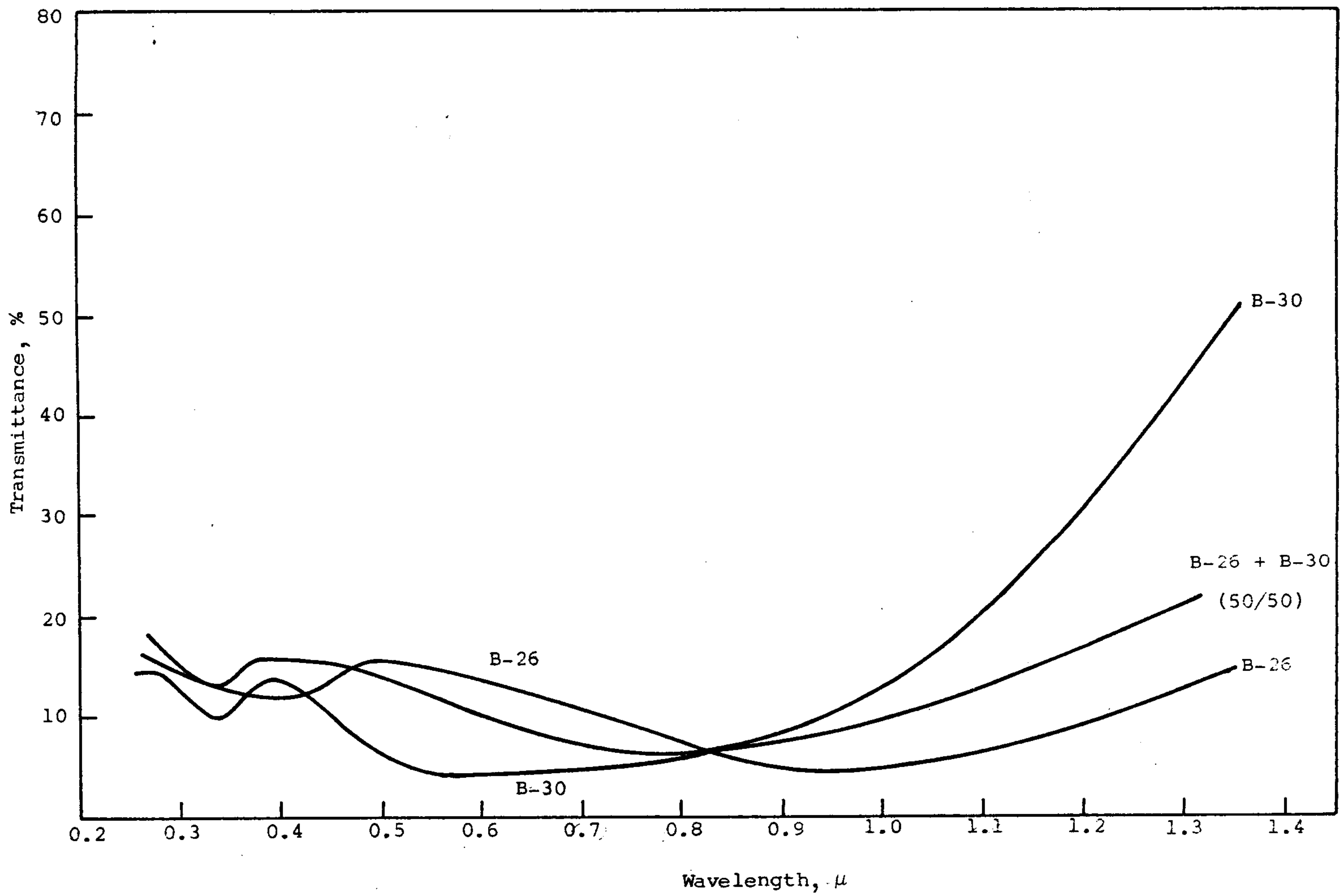

Figure 21

TRANSMITTANCE OF DILUTE SUSPENSIONS OF SILVER BROMIDE MIXTURES (Batches 26 and 30)

Thus the optical densities would be expected to be additive for each single energy loss mechanism, e.g., scattering (transmission equation) or absorption (Beer's law). At shorter wavelengths both these mechanisms apparently prevail.

A series of transmittance measurements were also obtained for mixtures of two particles sizes in concentrated suspensions. The results indicate that as in suspensions of single particle sizes, the particles in monolayers tend to act independently. But at film thicknesses corresponding to several layers, significant deviations occur. Additional quantitative data to support these observations will be given in future reports.

I. Secondary Oscillations in Scattering by Polystyrene Spheres

In addition to major oscillations in the total Mie scattering coefficients, Mie theory predicts a set of minor oscillations due to interferences of the reflected, refracted, and diffracted rays of the incident light. These are therefore consequences of the resonance of multipoles induced by the light wave.

Examination of the spectral transmittance curves obtained from suspensions of polystyrene spheres reveals a set of minor oscillations at the first scattering minimum which are very similar to the ones predicted by the theory.[3]

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[3] Penndorf, R.B., "Research on Aerosol Scattering in the Infrared," Final Report, Air Force Cambridge Research Laboratories, Technical Report RAD-TR-63-26 (Report AFCRL-63-668), 1963

The data in Figure 22 indicate that these oscillations (1) are not due to molecular absorption of polystyrene, (2) are independent of wavelength, and (3) are dependent on the effective refractive index. A shift in wavelength and relative positions can be easily noted by comparing the suspensions in water and in isopropanol. All these features strongly suggest that these oscillations are due to resonance of multipoles. A perfunctory search of the literature failed to yield any previous report of an actual experimental observation of these oscillations.

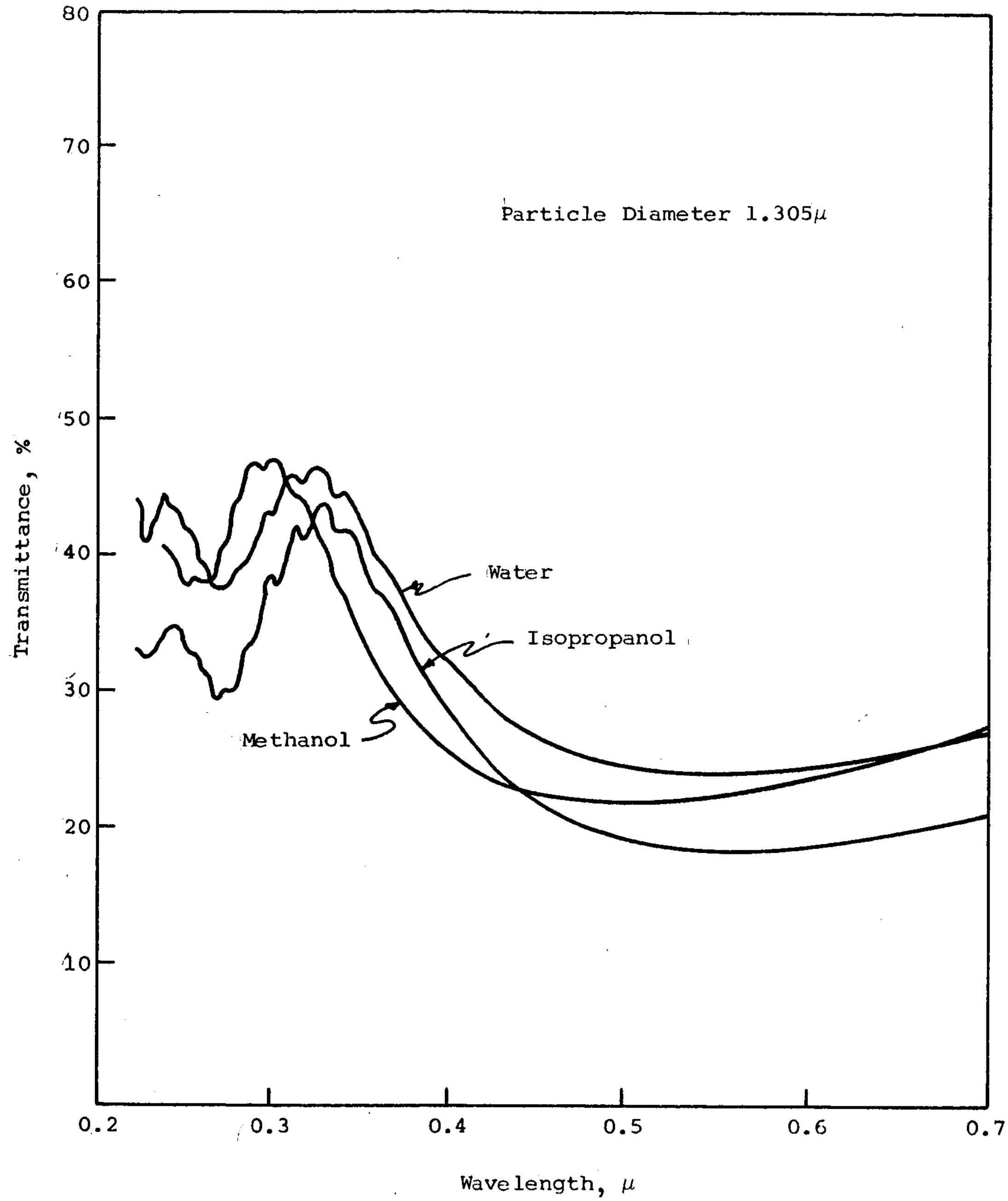

Figure 22

TRANSMITTANCE OF POLYSTYRENE SPHERES IN SUSPENSIONS OF VARYING REFRACTIVE INDEXES

IV. FUTURE WORK

Future investigations will emphasize continuing laboratory research on concentrated mono- and bimodal particle size distributions in gelatin films and theoretical analyses of multiple interactions in other than simple monodisperse, monolayer arrays. Other work will involve determination of radial distribution of total light scattering (unpolarized and plane polarized light) and back-scattering measurements.

Theoretical studies will involve increased emphasis on the literature search pertinent to particulate light scattering expecially as it relates to the multiple interaction of light in close-packed arrays of mono- and polydisperse systems.